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ARC STUDY OF THE OXYGEN SCHUMANN-RUNGE SYSTEM

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AYCO EVERETT RESEARCH LABORATORY

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ARC STUDY OF THE OXYGEN SCHUMANN-RUNGE SYSTEM^{*†}

by

Kurt L. Wray and Stephen Fried

December 1970

AVCO EVERETT RESEARCH LABORATORY
a division of
AVCO CORPORATION
Everett, Massachusetts

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FOREWORD

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This research was supported by the Advanced Research Projects Agency of the Department of Defense and Space and Missile Systems Organization, Air Force Systems Command and was monitored by Space and Missile Systems Organization, Air Force Systems Command under Contract F04701-70-C-0128. The work was initiated under a previous contract. The secondary report number as assigned by AERL is Avco Everett Research Laboratory Research Report 354. The Air Force program monitor for this contract is Capt. R. W. Padfield, USAF, Project Officer, Environmental Technology Branch, RNSE.

Publication of this report does not constitute Air Force approval of the report's findings or conclusions; it is published only for the exchange and stimulation of ideas.

Capt. R. W. Padfield
Project Officer,
Environmental Technology Branch,
RNSE

ABSTRACT

Other investigations have shown that the Schumann-Runge system of oxygen is excited by an inverse predissociation mechanism, $O + O \rightarrow O_2^*$. Under the condition where this excitation mechanism dominates, there is some question as to whether the radiation yielded a true rotational/vibrational electronic band spectrum, as opposed to a recombination continuum. Furthermore, there is a controversy as to whether the spectral intensity is that given by equilibrium theory. The present work utilized a 1 atm. arc jet facility to heat air and O_2 -noble gas mixtures to temperatures between 3000 and 3500°K. Both photographic and photoelectric spectra were recorded. In the latter case, wavelength scans from 2000 to 6000 Å were carried out with 5 Å resolution to obtain absolute intensity data. The spectra were found to have true rotational/vibrational electronic band structure, and were identified as the O_2 Schumann-Runge system. Comparison with an equilibrium radiation model showed excellent agreement. A slight adjustment was made in the electronic transition moment for the $NO(\beta)$ system as demanded by the air runs.

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1. INTRODUCTION

A recent shock tube study of the excitation mechanism for the O_2 Schumann-Runge (SR) system ($B^3\Sigma_u^- \rightarrow X^3\Sigma_g^-$) was carried out by one of the present authors¹. In that work the radiation behind the incident shock wave in O_2 and O_2 -noble gas mixtures was monitored as a function of time in two narrow bandpass channels centered at 3250 and 2300 Å. In the shock tube study the prime purpose was to ascertain the excitation mechanism for this system, although considerable information was also obtained on the spectral intensity at equilibrium. It was found, in agreement with the work of Myers and Bartle², that the radiating state ($B^3\Sigma_u^-$) was populated by inverse predissociation from ground state O-atoms, i.e., $O + O \rightarrow O_2^*$. However, the spectral distribution, and its temperature dependence, was found to be close to that given by equilibrium theory, in opposition to the results of Myers and Bartle who found large deviations from equilibrium spectral intensity.

The present work was undertaken principally to obtain high resolution spectra of the O_2 (SR) system. The motivation was two-fold: 1) to ascertain whether the spectrum was a true rotational/vibrational electronic spectrum as opposed to a continuum, as is found in many atom recombination processes and 2) to obtain continuous spectral intensity data over a sufficiently wide wavelength regime to ascertain whether the spectral distribution was that given by equilibrium theory. The experimental work reported in this paper was carried out employing a one atm. arc as the radiation source. Both air and O_2 -noble gas mixtures were used; the temperature range was $\sim 3000 - 3500^\circ K$. It should be pointed out that the oxygen concentration in the present arc studies is within the range of conditions covered in the shock tube studies^{1, 2}.

High resolution photographic spectra were obtained and these showed typical vibrational rotational electronic band structure. Spectral absolute intensities were obtained between 2000 and 6000 Å using a scanning double monochromator with photomultiplier detection. These intensities were compared with theoretical models including the O_2 (SR), NO(β) and (γ) systems and NO + O recombination continuum. Good agreement was found between experimental and equilibrium theory models for the Schumann-Runge spectral intensities. Furthermore, for the first time known to the authors, the NO(β) system was detected in high temperature air spectra, and has resulted in a revised electronic transition moment for this system.

II. EXPERIMENTAL TECHNIQUE

The arc facility³ used in this study runs at a pressure of one atmosphere and over a temperature range of 3000° to 5700°K (when the gas mixture is air). This facility has been used for extensive experiments on radiation from ablating boundary layers^{4, 5} as well as for studies of IR radiation from high temperature air^{6, 7}. The arc is struck across a flowing inert gas such as N₂, He or Ar which flows into a plenum chamber where it is mixed with oxygen. The hot gases exit into the atmosphere through a cylindrical constant area nozzle 1.9 cm in diameter and are viewed from the side with optical instruments. The equilibrium temperature of the gas is computed by subtracting the energy loss to the water-cooled walls (which is measured) from the total energy expended in the arc. For air, this computed energy balance temperature has an uncertainty of several hundred degrees and (measured) high frequency fluctuations of the order of $\pm 200^{\circ}\text{K}$.

An f/12 optical system, composed of a 4" mirror and two 90° flats, imaged the entrance slit of a 1/4 meter Jarrell-Ash (model #82-410) double monochromator at the centerline of the exiting arc heated gases, 2 mm downstream from the nozzle. Gratings blazed for 3000 Å were used in each monochromator over the region of interest which extended from 2000 to 6000 Å. The wavelength drives of each monochromator were synchronously driven by a belt supplied by the manufacturer for that purpose, and a cam and micro switch were set up to take off a wavelength marker signal every 25 Å. With the 100μ slits used in this study, the bandwidth was $\Delta\lambda = 5 \text{ Å}$ (full width at half height) and the scattered light less than 1 part in 10⁹. A

solar blind photomultiplier was used below 1100 \AA and 1P28 (with a 0-54 Corning filter to cut out second order) above 3000 \AA . Prior to entering the monochromator the light was mechanically chopped at 10^3 cps. The PM signal was electronically filtered through a narrow bandpass filter centered at the chopping frequency and then put onto a chart recorder along with the wavelength marker signal. The wavelength calibration for the system was obtained by scanning the output of a mercury lamp placed at the focus of the optics.

The optical system was calibrated for absolute intensity with a calibrated tungsten ribbon lamp. The system was found to be linear in response. The calibrations were made in two parts, scanning from 2000 \AA to 3100 \AA or 3000 \AA to 6000 \AA just before an arc run was to be made over the same wavelength interval. The calibration over the 2000 \AA to 3100 \AA interval was made possible by elimination of the scattered light problem by employing the double monochromator in conjunction with the solar blind P.M.

The spectral emissivity of tungsten is given by deVos⁸ for $\lambda \geq 2300 \text{ \AA}$. Recent work by Kostkowski⁹ at the National Bureau of Standards has given the emissivity of tungsten from $2000 - 2800 \text{ \AA}$ in the form of spectral radiance data (includes quartz window material) for a source identical to that used in the present work. These data were in good agreement with those of de Vos⁸ at 2800 \AA and were $\approx 25\%$ lower at 2300 \AA . The emissivities given by Kostkowski⁹ were used in the present work for $\lambda \leq 2800 \text{ \AA}$.

A typical example of the results of this calibration is shown in Fig. 1 where the sensitivity in $\text{mv/watt /cm}^2\text{-ster-\mu}$ is plotted against wavelength for the two regimes discussed above. Note that because of the extremely steep wavelength dependence between 3000 and 3100 Å for both detectors that some inconsistencies in the data might develop in this region of spectral overlap.

Prior to taking the quantitative spectral data with the scanning instrument, qualitative data, in the form of photographic spectra, were obtained. For this purpose a f/10 Hilger prism spectrograph with quartz optics was used employing a 15μ slit and Kodak 103f film.

Runs were made in air (21% O_2 - 79% N_2 mixtures) at temperatures of ~ 3200 and $\sim 3500^\circ\text{K}$ and in several O_2 -He and O_2 -Ar mixtures. When running on either He or Ar, the arc efficiency drops drastically so that the uncertainty in the gas enthalpy increases significantly. Hence, for these cases, the uncertainty in the gas temperature is somewhat greater than in the air case.

All of the intensity vs. wavelength data were digitized by reading on an oscar, processed on an IBM 360-44 computer, and machine plotted.

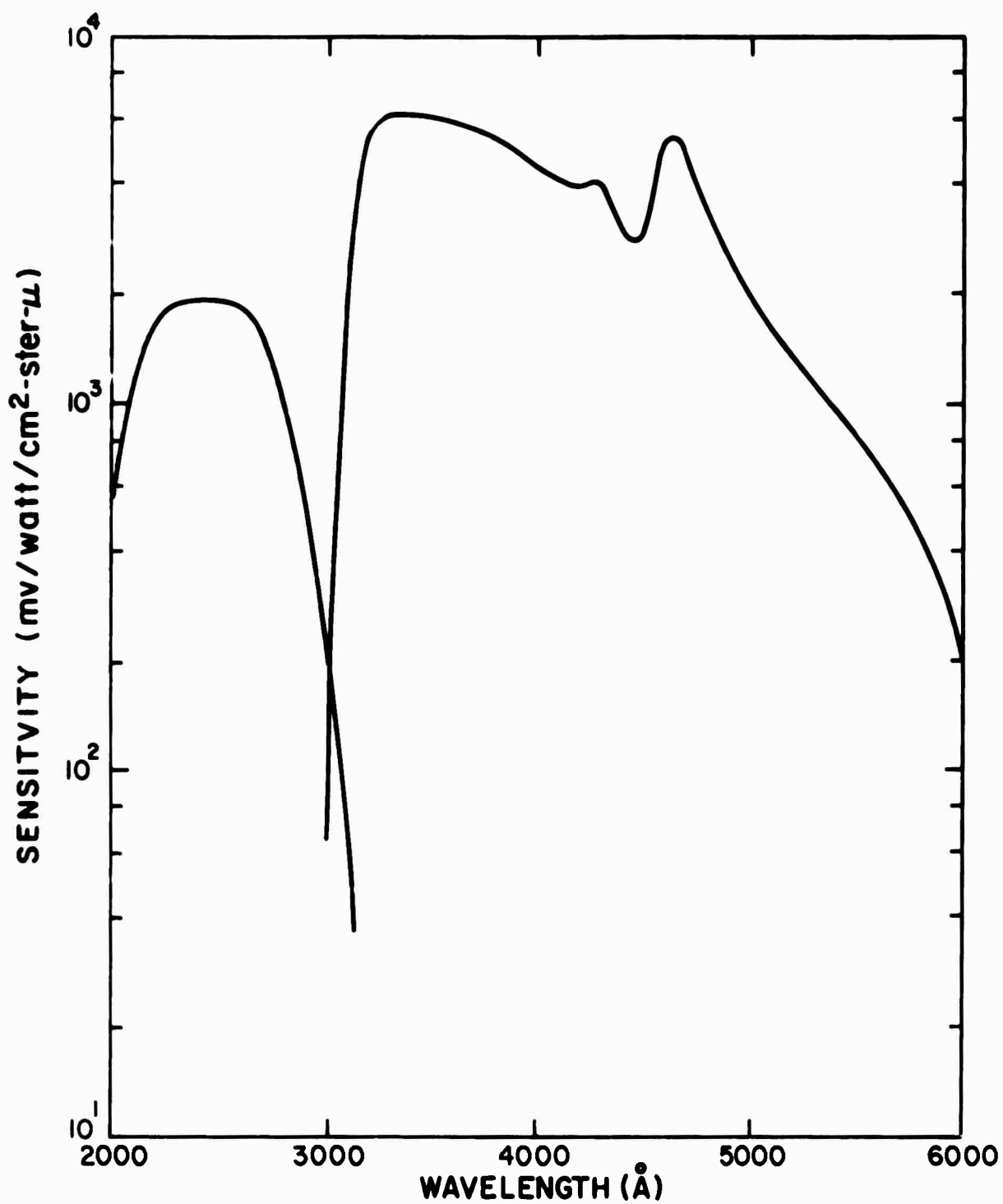


Fig. 1 Calibration curves for the optical system. The system covering 2000 - 3100 Å employed a solar blind photomultiplier.

III. CALCULATION OF THEORETICAL SPECTRA

In this section the models used to calculate the theoretical spectra are briefly described. It was assumed in all cases that the gas was optically thin. All models, of course, require the concentration of radiating species. These were obtained at any required temperature utilizing our Thermo-Chemical Program employing the latest JANNAF¹⁰ thermodynamic properties.

A. O₂ (S.R.)

The spectral intensity of the ($B^3\Sigma_u^- \rightarrow X^3\Sigma_g^-$) system was computed line by line using a model similar to that previously described⁶. The nature of the model can be indicated to the interested reader by noting that the spectroscopic constants were given by the well-known relations:

$$F_{v,J} = B_v J(J+1) \quad ,$$

$$B_v = B_e - \alpha_e (v + 1/2) \quad ,$$

$$G(v) = \omega_e (v+1/2) - \omega_e x_e (v+1/2)^2 + \omega_e y_e (v+1/2)^3 + \omega_e z_e (v+1/2)^4 \quad ,$$

where all the notation is conventional. The constants used were those of Hebert, Innanen and Nicholls¹¹. The electronic transition moment $\left| \frac{R(r_e)}{ea_0} \right|^2$ is a function of the r-centroid, r_e , and it varies from band to band. Marr¹² has determined a linear fit to experimental values of $\left| \frac{R(r_e)}{ea_0} \right|^2$ vs. r_e which was used in conjunction with the r-centroids of Hebert, et al¹¹. The Franck-Condon factors of Ory and Gittleman¹³ were used for $v' \geq 2$ which took into account the asymmetries of the upper state; for $v' = 0$ and 1 those of Hebert, et al.¹¹ were used.

The fine structure due to spin coupling was averaged over by simplifying the Hönl-London factors $S_{J'}$ to yield only an R and P branch whose line strengths were given by $S_{J'} = J'$ and $S_{J'} = J' + 1$, respectively, where J' is the rotational level in the upper state. Because the oxygen nucleus has zero spin angular momentum, only the odd numbered rotational transitions appear. A total of 196 vibrational bands were utilized in the computation which spanned the spectral region of 1900 to 6000 Å; rotational states up to $J = 200$ were included. To simulate the response and resolution of the spectrometer a Gaussian transmission function was utilized to average the output lines. It was found that an 8 Å Gaussian half width positioned at 2 Å intervals reproduced the instrumental response.

B. NO(β) and NO(γ) Systems

It is well known¹⁴ that NO band systems contribute to air radiation in the portion of the spectrum investigated in the present work. These include both the NO(β) ($B^2\pi \rightarrow X^2\pi$) and the NO(γ) ($A^2\Sigma^+ \rightarrow X^2\pi$) systems. The theoretical computation of the spectral intensities for these two systems employed the smeared rotational model (and q-branch approximation)^{14, 15, 16} and is a direct descendent of the computer program previously employed by Allen¹⁷. The electronic transition moment $\left| \frac{R(r_e)}{ea_0} \right|^2$ for the γ system was taken to be 0.025¹⁷ while that for the β system was adjusted down from 0.08¹⁷ to 0.04 as a result of the present investigation.

C. NO + O \rightarrow NO₂ + $h\nu$

It is necessary to include the NO + O chemiluminescence radiation to explain the near U. V. behavior of the experimental data. This process

has been investigated by Fontijn, Meyer and Schiff¹⁸; their results give the intensity I in a wavelength region $\lambda_1 \leq \lambda \leq \lambda_2$ as

$$I = \frac{[O][NO]k}{4\pi(\lambda_2 - \lambda_1)} \frac{\int_{\lambda_1}^{\lambda_2} I_{\lambda} d\lambda}{\int_{3,875}^{14,000} I_{\lambda} d\lambda} \frac{hc}{(\lambda_2 + \lambda_1)/2}$$

where $k = 6.4 \times 10^{-17} \text{ cm}^3 \text{ molecule}^{-1} \text{ sec}^{-1}$. The function in the integral is given in tabular form in Ref. 18.

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IV. EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

Table I lists the composition, temperature and wavelength regimes covered for the runs relevant to this paper. Some air runs at higher temperatures were made, but the spectral features were dominated by the $N_2^+(1-)$ and $N_2(2+)$ systems and hence are not relevant to the present paper. All of the spectra listed in Table I show near identical features although the absolute intensity varies depending on the temperature. Because of this similarity between spectra not all of the experimental data is reproduced here in detail.

In Fig. 2 are shown two examples of the photographic spectra obtained with the Hilger instrument. The wavelength regime covered was limited on the blue side to $\sim 2500 \text{ \AA}$ and on the red side to $\sim 7000 \text{ \AA}$ by the sensitivity of the 103 f film. Spectrum A (Run #1) is a 2 minute exposure on a run made in a 16% O_2 -84% He mixture. The energy balance temperature for the oxygen-noble gas mixtures is somewhat uncertain but is within several hundred degrees of 3500°K . Spectrum B (Run #2) is a 5 minute exposure made on an air run with $T = 3100^\circ\text{K}$. The two spectra obviously show a great deal in common; the bands whose intensity on the photographic plate peak at about 3000 \AA are identical and can be matched rotational line for line.

If there is a continuum behind this rotational/vibrational electronic spectrum, it is relatively weak. The air spectrum (B) does show a strong continuum developing toward the red portion of the spectrum. This is completely missing in the absence of nitrogen as seen in Spectrum A. As will be shown this continuum is due to radiative recombination of O and NO.

TABLE I
RUN CONDITIONS FOR AIR AND O₂-NOBLE GAS MIXTURES

<u>Run #</u>	<u>Composition (%)</u>	<u>T (°K)</u>	<u>λ (Å)</u>
1	16 O ₂ - 84 He	3000 - 3500	103 f film
2	21 O ₂ - 79 N ₂	3100	103 f film
3*	16 O ₂ - 84 He	3000 - 3500	3100 → 4000
4*	16 O ₂ - 84 He	3000 - 3500	3200 → 4000
5	21 O ₂ - 79 N ₂	3155	2000 → 3100
6	21 O ₂ - 79 N ₂	3520	2000 → 3100
7	21 O ₂ - 79 N ₂	3130	3000 → 6000
8	21 O ₂ - 79 N ₂	3265	3000 → 6000
9	21 O ₂ - 79 N ₂	3480	3000 → 6000
10	21 O ₂ - 79 N ₂	3590	3000 → 4200
11	21 O ₂ - 79 N ₂	3630	3000 → 3600
12	16 O ₂ - 84 He	3000 - 3500	2000 → 3100
13	7.8 O ₂ - 92.2 Ar	3000 - 3500	2000 → 3100

* = low resolution runs.

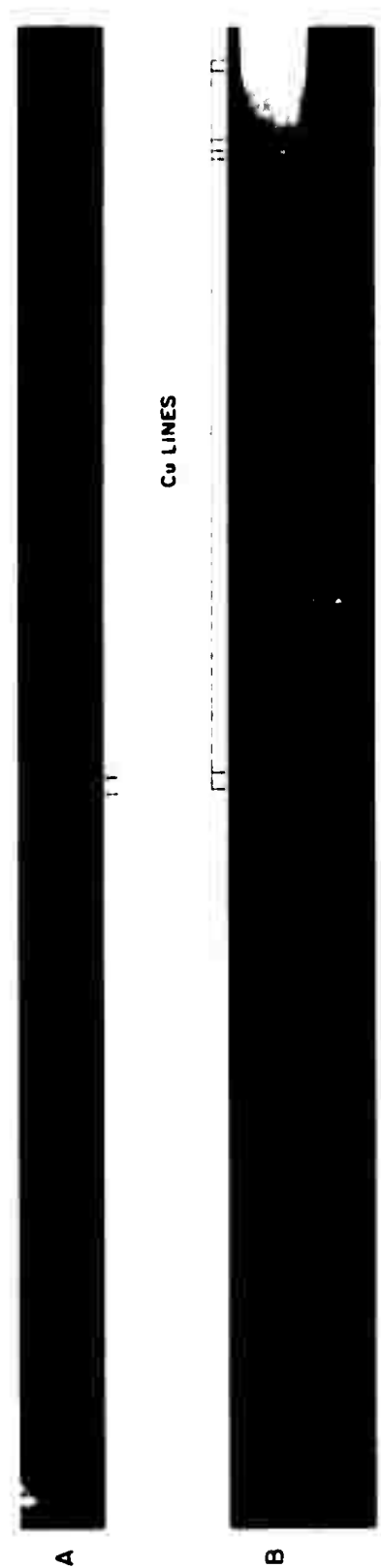


Fig. 2 Photographic spectra obtained with an f/10 Hilger prism spectrograph with quartz optics, 15μ slit and Kodak 103f film. The copper lines are present as a trace impurity in the arc heated gases.

A. 16% O_2 - 84% He, exposure = 2 min, $3000 \lesssim T \lesssim 3500^\circ K$.

B. Air, exposure = 5 min, $T = 3100^\circ K$.

The only impurities observed in the spectra are the copper lines which are identified in the figure. Earlier measurements have shown copper to be present in the arc heated gases at about 10 PPM in the temperature regime covered in the present work. The rotational line spectra shown in Fig. 2A and B have been compared in detail with published SR spectra excited by a variety of techniques. There is no doubt concerning the identification of this radiation.

In Fig. 3 A and B (Runs #9 and 10) are shown portions of unreduced spectral scan data; hence this is relative intensity vs. wavelength. The copper lines at 3248 and 3374 Å are identified along with the position on the traces where gain changes were made during the run so as not to confuse these with spectral features. Two separate runs are shown in A and B demonstrating the high degree of reproducibility of the data. For comparison, Fig. 3C shows a theoretically computed O₂(SR) spectrum. Careful inspection of the theoretically computed spectrum based on the model described in Section III again identifies the experimental spectrum as O₂(SR).

Records such as are shown in Fig. 3 A and B can be converted into absolute intensity using calibration curves like that shown in Fig. 1. In Fig. 4 are shown the experimental results (Runs #3, 4, 12 and 13) for several of the O₂-noble gas runs in the 2000 - 4000 Å wavelength regime which is most critical for verifying the spectral intensity distribution. The experimental curves for 16% O₂ which go out to 4000 Å are poor resolution Runs #3 and 4. These were taken as preliminary data in the present investigation; however, due to difficulty in making noble gas runs, they were never redone with the 5 Å resolution ultimately achieved. However,

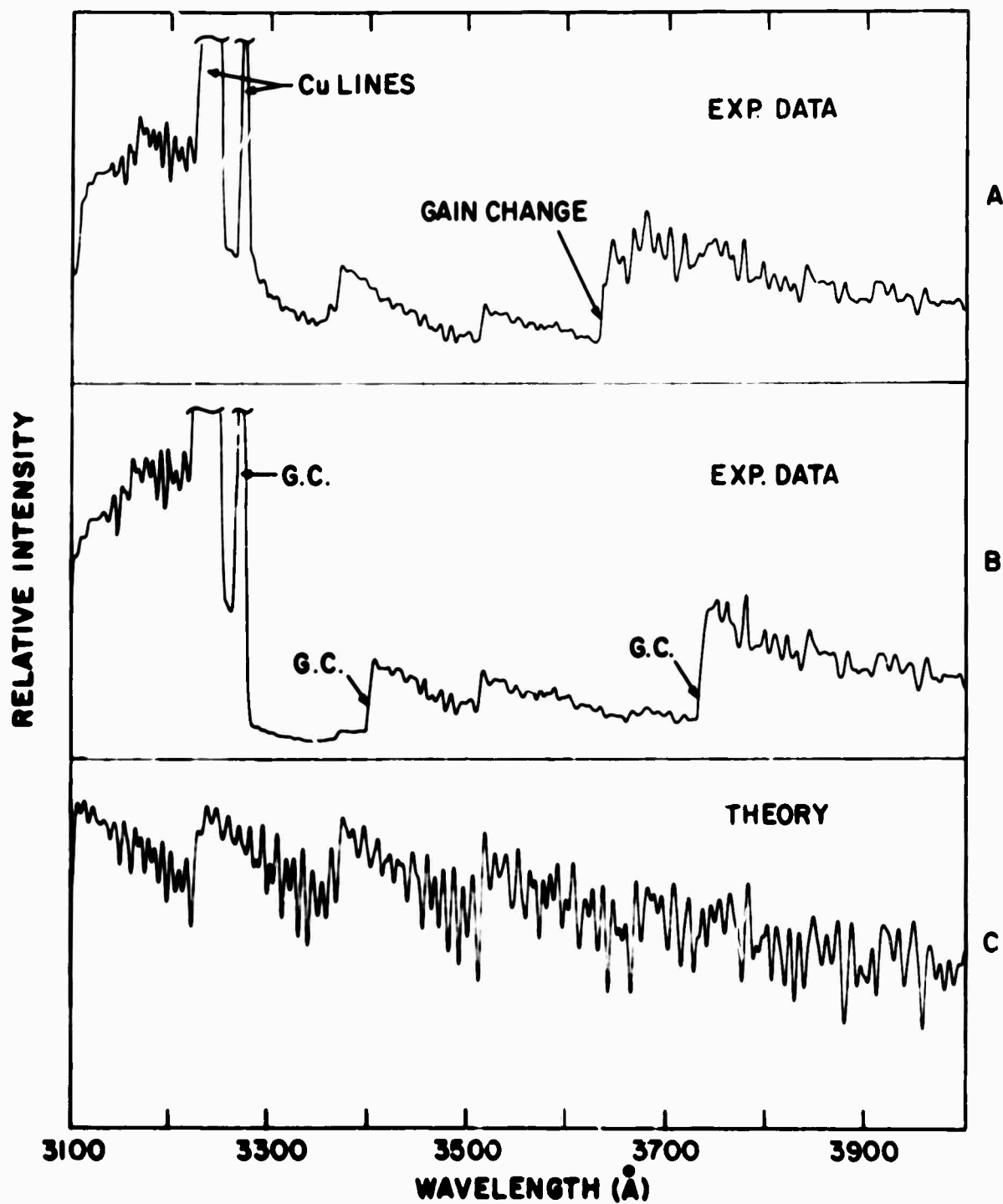


Fig. 3 A. Portion of chart recorder trace Air at $T = 3480^{\circ}\text{K}$.
 B. Portion of chart recorder trace. Air at $T = 3590^{\circ}\text{K}$.
 C. Theoretically computed O_2 (SR) spectrum ($T = 3500^{\circ}\text{K}$).

the detailed band structure shown in such bands as the (0, 12) (0, 13) (0, 14) (0, 15) out to the (1, 18) again verifies this radiation as being O_2 (SR) and for this reason we include these low resolution data.

Also shown in Fig. 4 for comparison with the experimental curves is a theoretical O_2 (SR) calculation for 16% O_2 at $3500^\circ K$. This curve is multiplied by a factor of 10 in Fig. 4 so as to prevent confusion by its overlap with the experimental data. The agreement between equilibrium theory and experiment is excellent both in terms of detailed structure and spectral intensity.

The air data nominally at 3200 and $3500^\circ K$ and covering the entire wavelength regime from 2000 to 6000 \AA are plotted in Fig. 5 (Runs # 5, 6, 8 and 9 in Table I.) Again the high degree of reproducibility between runs is apparent. The presence of the continuum at longer wavelengths is clear, as it was in the photographic spectrum of Fig. 2B. For comparison with the data shown in Fig. 5, theoretical radiation curves have been produced from the models discussed in Section III for a temperature of $3500^\circ K$; these are shown in Fig. 6. The three curves shown are for the contributions due to $NO(\beta)$, $NO(\gamma)$, and O_2 (SR) to which has been added the $NO + O \rightarrow NO_2 + h\nu$ radiation. These curves should be compared directly with the $3500^\circ K$ experimental data shown in Fig. 5.

As can be seen the theoretical model indicates that between 2000 and 2500 \AA the structure is dominated by the $NO(\gamma)$ system although O_2 (SR) radiation adds significantly between the $NO(\gamma)$ peaks. Between 2500 and 4500 \AA the SR radiation dominates the spectrum with the $NO(\beta)$ system contributing $\sim 1/3$ the radiation intensity. In the calculations of Fig. 6

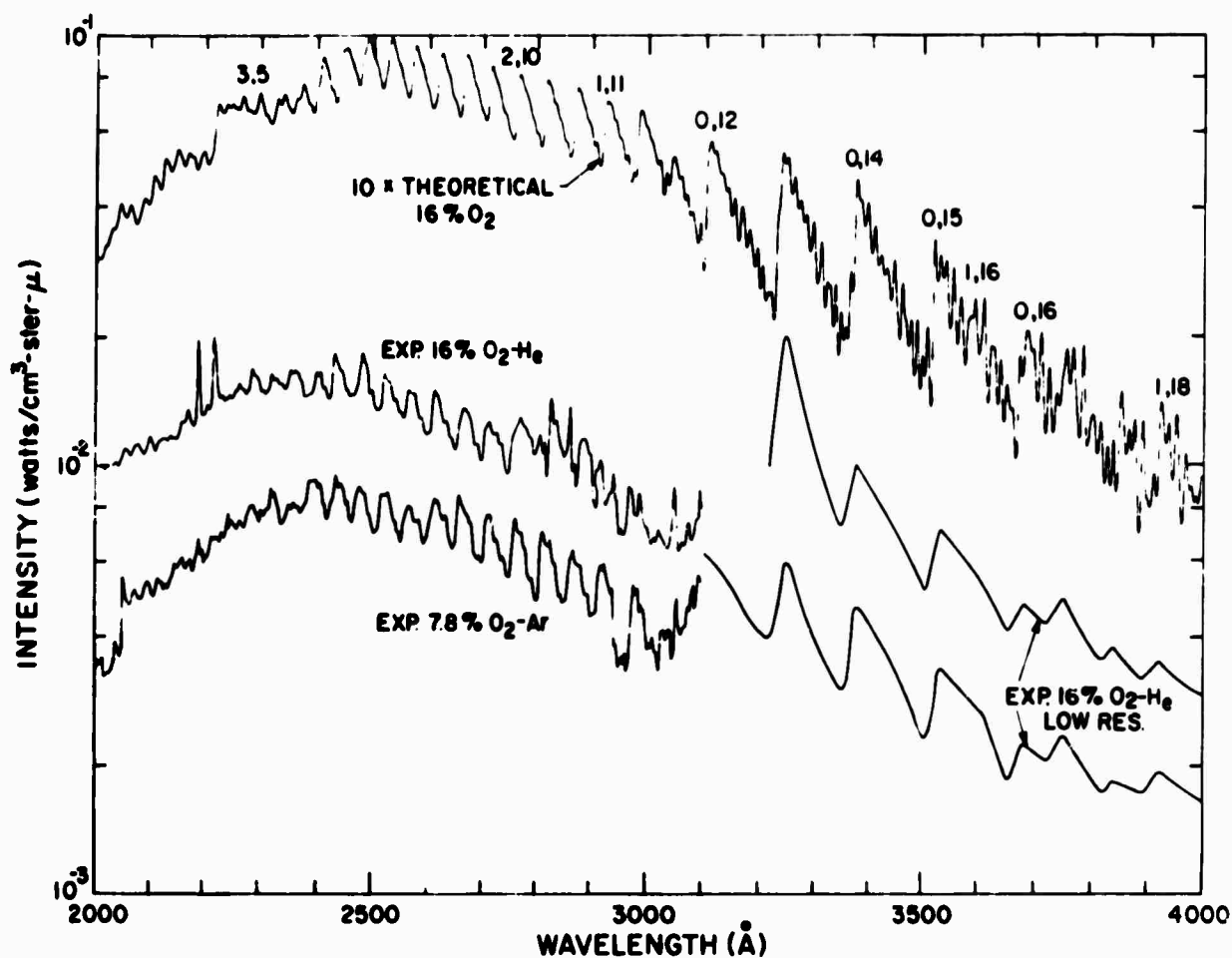


Fig. 4 Absolute intensity versus wavelength for a number of O_2 -noble gas runs ($3000 \leq T \leq 3500^\circ K$) compared with a theoretical calculation ($T = 3500^\circ K$). The data above 3100 \AA was taken under low resolution conditions. Note that the theoretical curve has been multiplied by a factor of 10 to prevent overlap with the data.

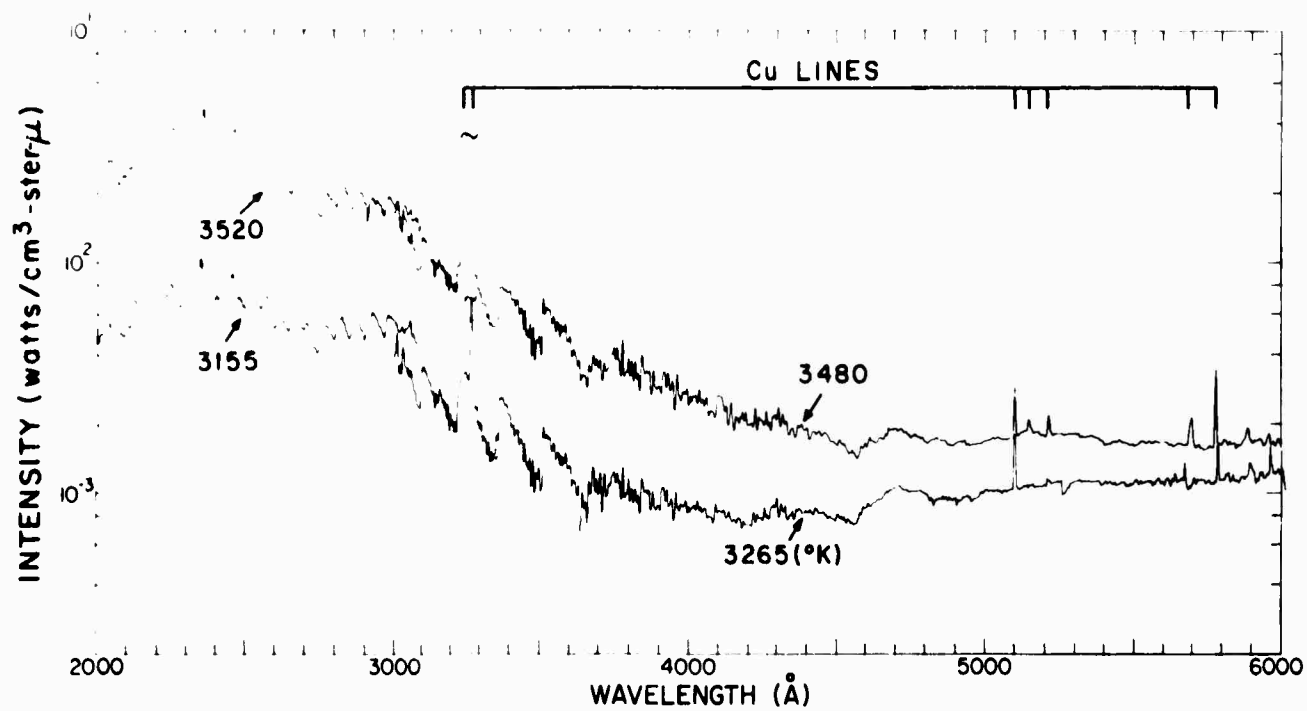


Fig. 5 Absolute intensity of a number of air runs plotted against wavelength.

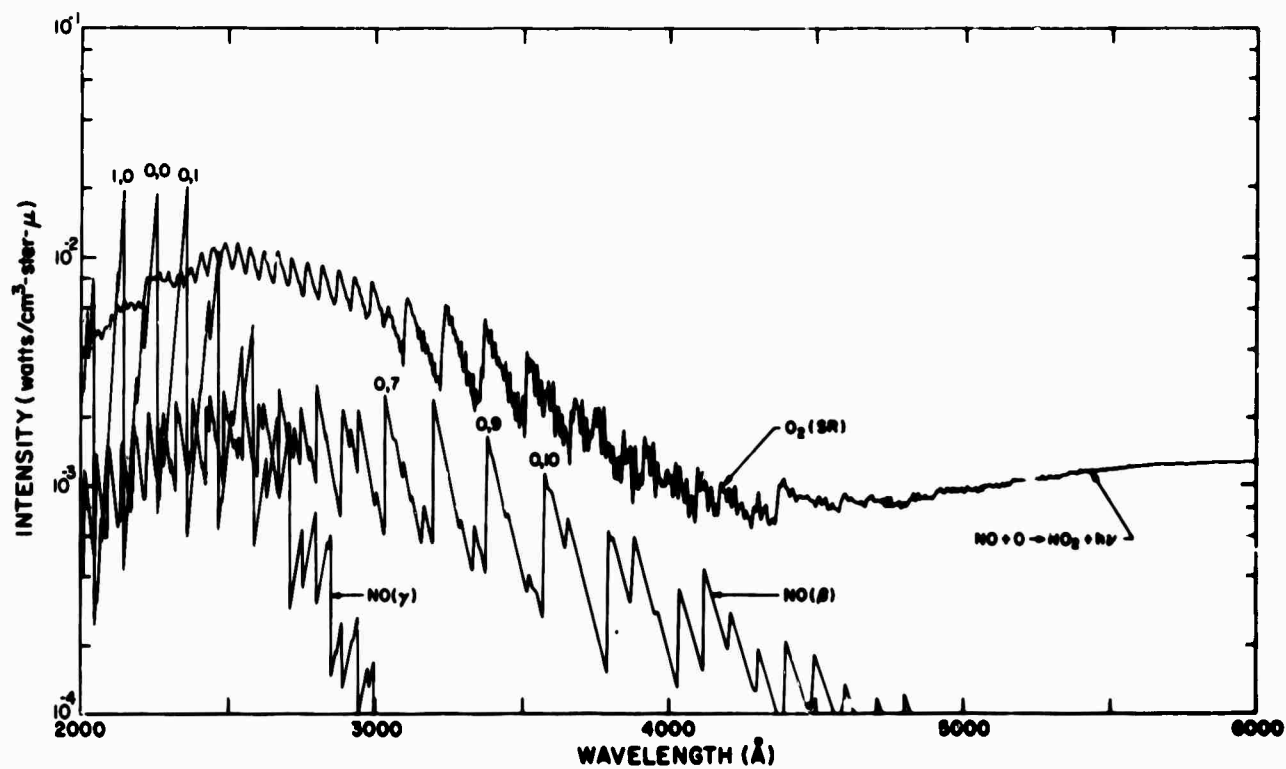


Fig. 6 Absolute intensity versus wavelength for the theoretically computed radiations. These curves should be compared with the data of Fig. 5.

the electronic transition moment for the NO(δ) system was reduced by a factor of two from that used at the onset of this work. This adjustment was made so as to fit some of the detail in the 3500 to 4000 Å regime where structure due to both the O₂(SR) and NO(δ) systems is apparent in the experimental data.

V. SUMMARY AND CONCLUSIONS

From the photographic spectra shown in Fig. 2 it has been demonstrated that the radiation is indeed a rotational/vibrational electronic spectrum and that the radiation obtained for arc heated air is similar in detailed structure to that for arc heated O_2 -noble gas mixtures. This rotational line spectrum has been identified as the $O_2(SR)$ system as excited by a variety of sources.

The scanning spectrometer yielded spectral intensity data which were in good agreement with theoretical equilibrium conditions. This finding is, within experimental error, in agreement with the recently completed shock tube study¹ which monitored the radiation in narrow bands at 3250 and 2300 Å and is in disagreement with the spectral distribution found in the shock tube study of Myers and Bartle², whose results indicated the intensity in the 2300 Å channel to be two orders of magnitude less than that given by equilibrium theory. It should be again pointed out that all three of these investigations covered similar atomic oxygen concentrations and similar temperature regimes. It must therefore be concluded that the spectral distribution reported by Myers and Bartle is in error. Hence, the inverse predissociation mechanism by which the SR state is populated is sufficiently fast under the experimental conditions of these investigations that all (thermally significant) vibrational and rotational levels of the radiating state ($B^3\Sigma_u^-$) have their equilibrium population. This equilibrium between all vibrational and rotational states of the B state is probably accomplished via the mechanism $O + O \rightleftharpoons O_2^*$ rather than selective population through this mechanism followed by collisional redistribution in the B state.

VI. ACKNOWLEDGMENTS

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13. ABSTRACT Other investigations have shown that the Schumann-Runge system of oxygen is excited by an inverse predissociation mechanism, $O + O \rightarrow O_2^*$. Under the condition where this excitation mechanism dominates, there is some question as to whether the radiation yielded a true rotational/vibrational electronic band spectrum, as opposed to a recombination continuum. Furthermore, there is a controversy as to whether the spectral intensity is that given by equilibrium theory. The present work utilized a 1 atm arc jet facility to heat air and O_2 -noble gas mixtures to temperatures between 3000 and 3500°K. Both photographic and photoelectric spectra were recorded. In the latter case, wavelength scans from 2000 to 6000 Å were carried out with 5 Å resolution to obtain absolute intensity data. The spectra were found to have true rotational/vibrational electronic band structure, and were identified as the O_2 Schumann-Runge system. Comparison with an equilibrium radiation model showed excellent agreement. A slight adjustment was made in the electronic transition moment for the $NO(\beta)$ system as demanded by the air runs.			

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KEY WORDS

1. O₂ Schumann-Runge radiation
2. High temperature air radiation
3. Arc heated gases
4. Electronic transition moments
5. NO (β) system
6. NO (γ) system
7. NO + O recombination continuum

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